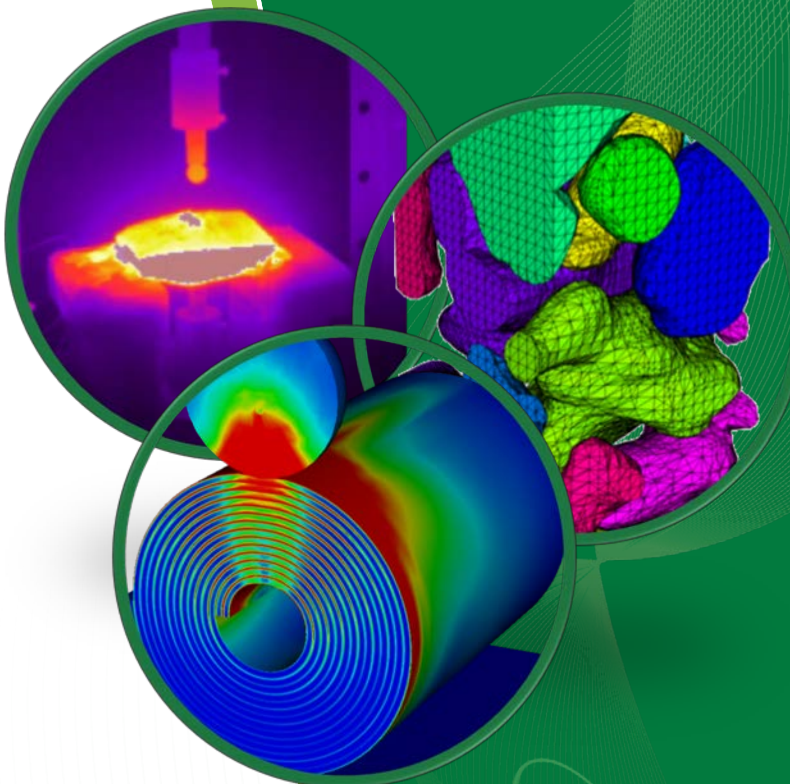
Exploring How Electrode Structure Affects Electrode-Scale Properties Using 3-D Mesoscale Simulations



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Organization: Sandia National Laboratories

Team: Consortium for Advanced Battery Simulation

2018 U.S. DOE Vehicle Technologies Office Annual Merit Review

June 20, 2018 Project ID: BAT303

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CABS Consortium for Advanced Battery Simulation

Oak Ridge National Laboratory · Argonne National Laboratory · Berkeley Lab · Sandia National Laboratories

Overview

Timeline

- Project Start Date: Oct 1, 2015
- Project End Date: Sept 30, 2018
- Percent Complete: 92%

Barriers Addressed

- **Life:** Loss of available power and energy due to use and aging, and the lack of accurate life prediction capability.
- **Abuse Tolerance, Reliability and Ruggedness:** It is critical that any new technology introduced into a vehicle be abuse tolerant under both routine and extreme operating conditions.

Budget

- FY17
  - Total CABS: $2,225k
  - SNL Effort: $500k
- FY17
  - Total CABS: $1,398k
  - SNL Effort: $450k

Partners

- Project Partners/Consortium:
  - Oak Ridge National Laboratory
  - Lawrence Berkeley National Laboratory
  - Argonne National Laboratory
- NREL-led CAEBAT team
  - SNL, TAMU

Relevance / Objectives

- Project Objectives
  - Improve the fidelity of battery-scale simulations of abuse scenarios through the creation and application of microscale (particle-scale) electrode simulations
- Present Year Objectives
  - Perform coupled mechanical-electrochemical discharge simulations in normal and abuse environment
  - Model electrode swelling/breathing
- Impact to VTO
  - Improve ability to assess battery response to abuse scenarios (e.g. crush) computationally, enabling many parametric computer tests rather than expensive and dangerous experiments

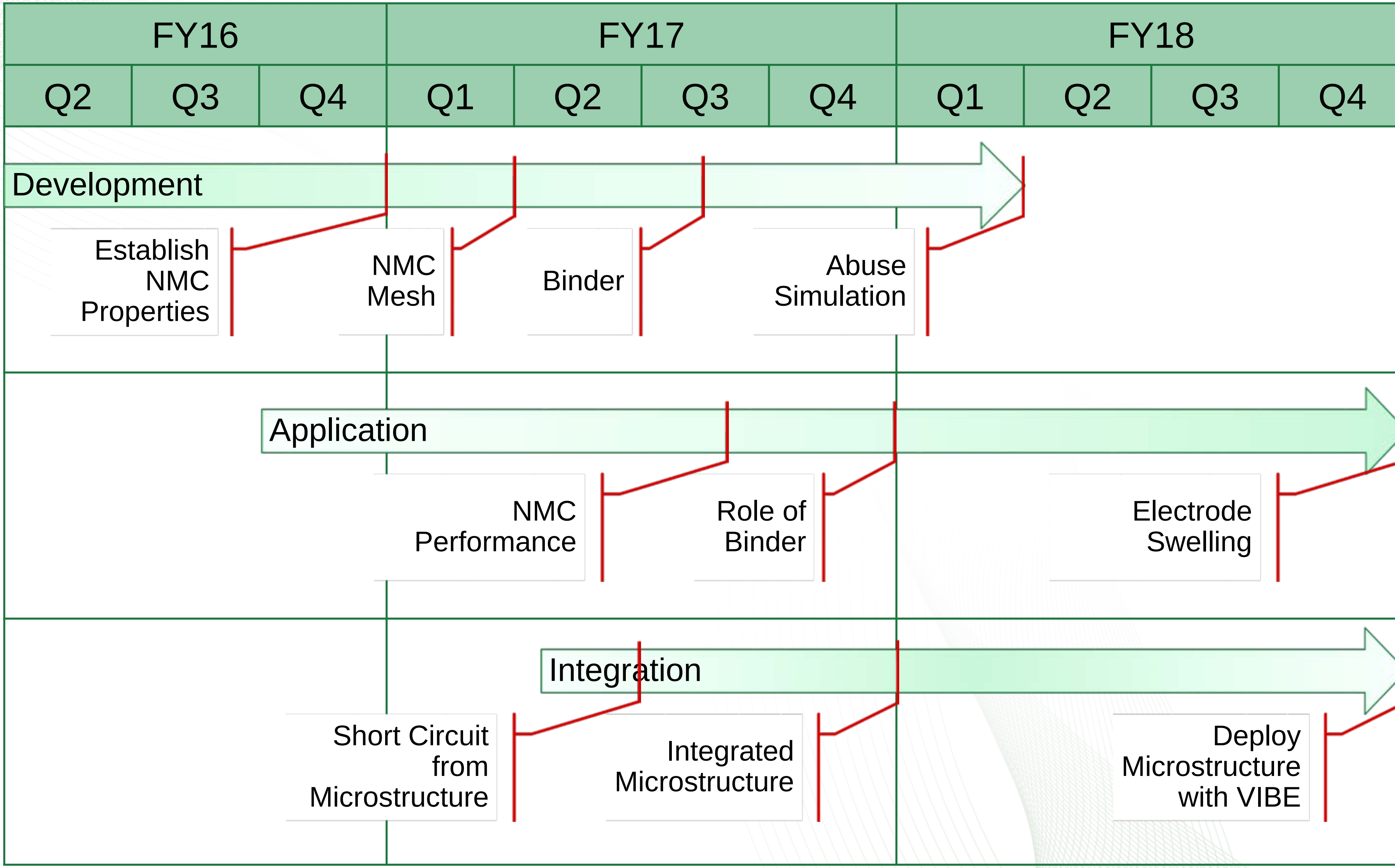
CABS Milestones (FY17)

IDs indicate whether milestones are primarily experimental (E), computational (C), or integrated (I).							
ID	FY17	Lead	Q1	Q2	Q3	Q4	Status
I.3	***Demonstration of ability to construct 3D meshes of electrodes using reconstructions from micro-tomography***	***SNL***	***P***				***Complete***
E.3	Potential-dependent solid diffusivities for Li-ion and EIS	LBNL		P			Complete
I.4	*Demonstrated ability of VIBE/OAS to simulate onset of short-circuit due to mechanical abuse informed by microstructure*	*ORNL*		*D*			*Complete*
E.4	Data from mechanical deformation tests	ORNL			P		Complete
C.2	Validated constitutive models and failure criteria for electrode materials and spirally wound, wound prismatic, and stacked electrodes under indentation	ORNL				P	Ongoing
I.5	*Deployment of VIBE/OAS with integrated multiscale capability*	*ORNL*				S	*Ongoing*

CABS Milestones (FY18)

IDs indicate whether milestones are primarily experimental (E), computational (C), or integrated (I).							
ID	FY17	Lead	Q1	Q2	Q3	Q4	Status
C.3	Coupled thermo-electro-mechanical microstructure simulations of overcharge and mechanical abuse scenarios	***SNL***	***P***				***Ongoing***
E.5	Obtain electrode image data from cycled electrode material	LBNL	P				Ongoing
C.4	Demonstrated mesoscale simulations	ORNL		P			Ongoing
C.5	Demonstrate improved computational efficiency on a benchmark pack-level simulation using a hierarchy of electrochemical models for US06 drive	ORNL			P		Ongoing
C.6	Validated constitutive models & failure criteria for electrode materials & spirally wound, wound prismatic, & stacked electrodes under bending for pouch cell	ORNL				P	Ongoing
I.5	*Deployment of VIBE/OAS with efficient, validated mechanistic models*	*ORNL*				S	*Ongoing*

Approach / Milestones

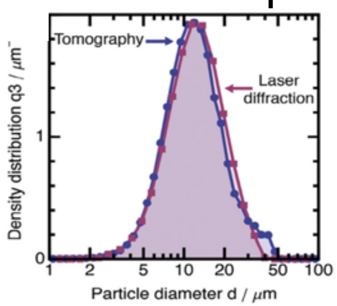


Technical: Electrode Manufacturing with DEM

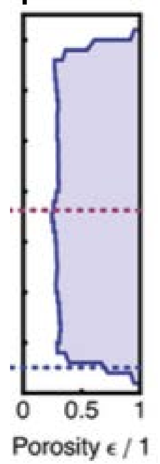
- Use Discrete Element Method (DEM) simulations to:
  - Predict mesostructure independent of tomography
  - Improve understanding of how manufacturing conditions (e.g. mixing, deposition, coating, drying, and calendering) affect mesostructure
- DEM includes contact mechanics, friction, cohesion, and drag

Unknown cohesive surface energy calibrated to experimental data

Use particle size from expt.

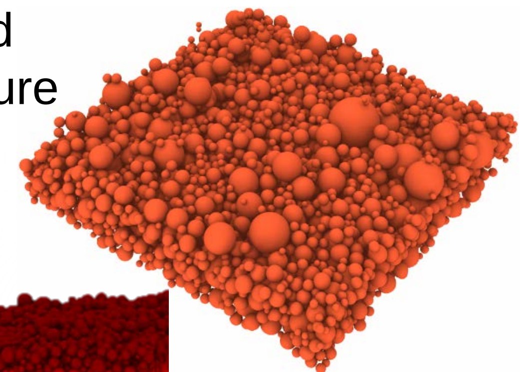


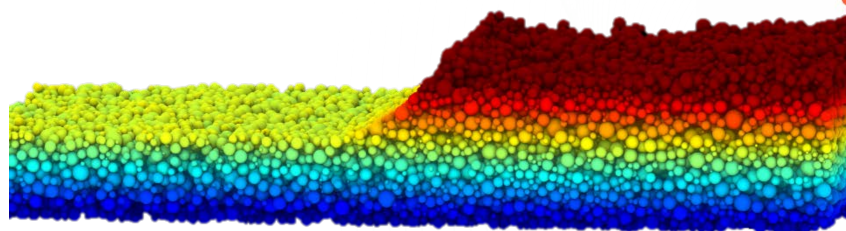
Ebner (2013)



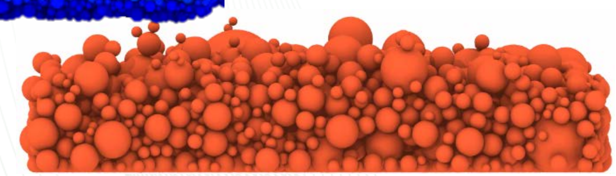
Ebner (2013)

Simulated mesostructure





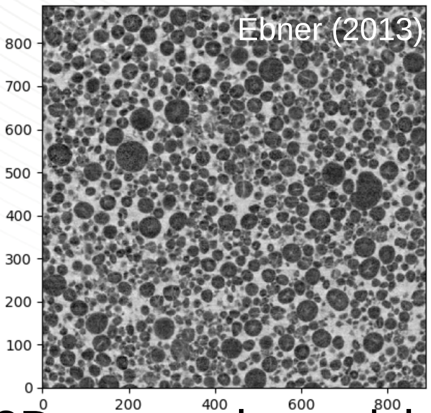
Calendering



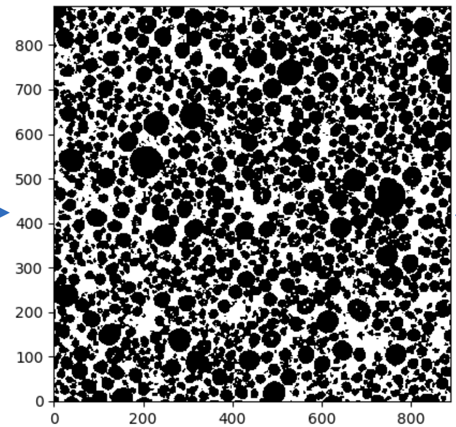
Validated DEM simulations to provide manufacturing insight

Technical: Image to Mesh with Three Phases

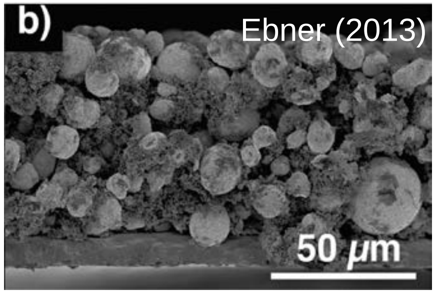
- Enhancing workflow to convert 3D tomography to mesh



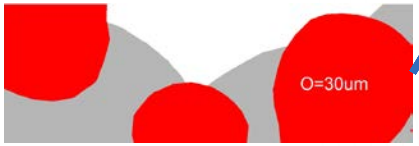
3D greyscale particles



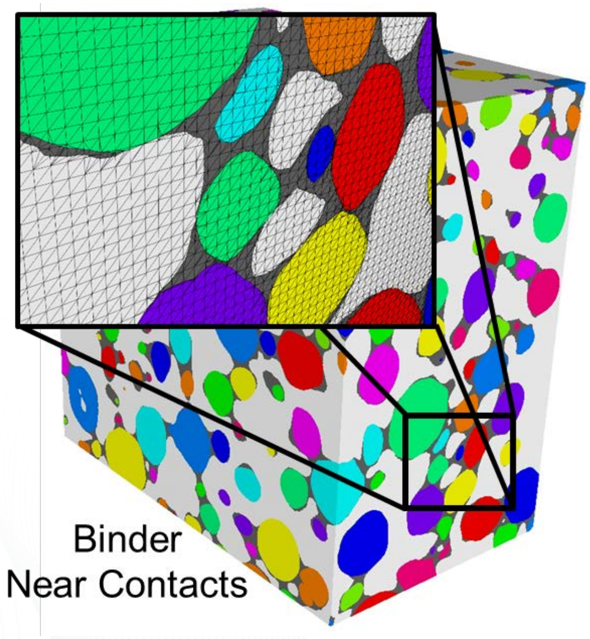
Machine learning segmentation



2D conductive binder images - motivation



"Binder bridge" morphology generation



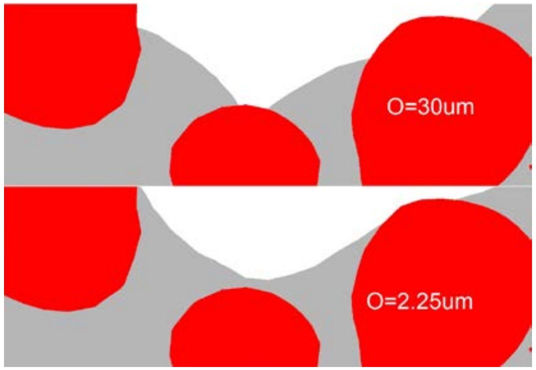
CDFEM-generated 3D, 3 phase mesh

Efficient method for creating high-quality mesoscale mesh

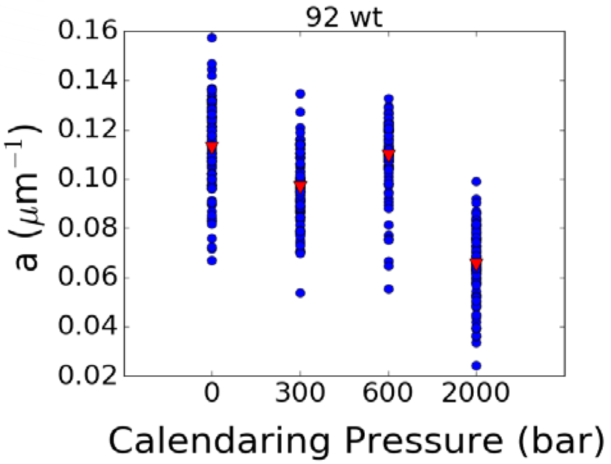
Roberts et al (2014, 2016)
Trembacki et al (2017)

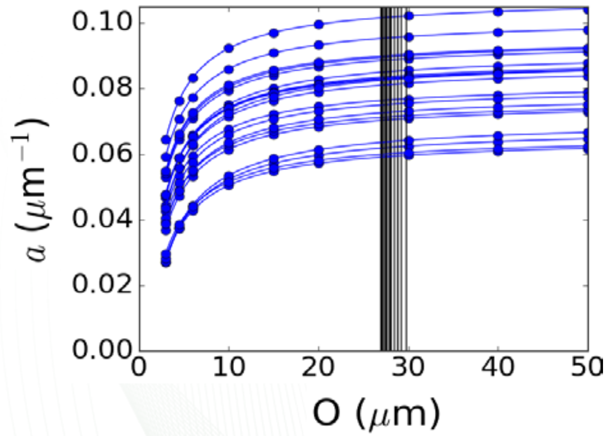
Technical: CBD Morphology

- Choice of CBD parameters influences morphology



- Large subdomain-subdomain variability in particle density requires statistical analysis of hundreds of subdomains





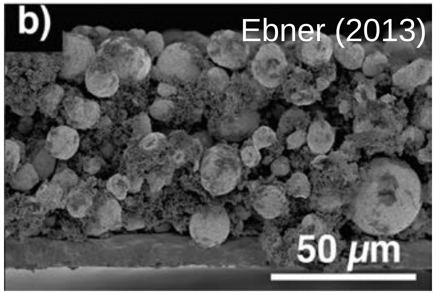
- Parameters statistically optimized to get correct CBD loading

Focusing on getting the CBD domain right

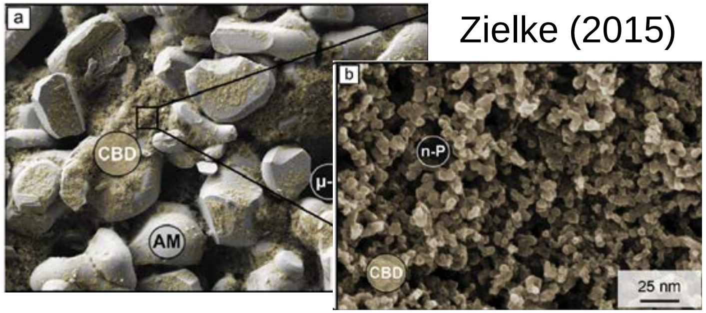
Trembacki et al, in preparation

Technical: Nanoporosity in CBD

- Imaging suggests carbon binder domain (CBD) ~ 50% porous
  - Can lead to as much as a 0.40:1 CBD:particle volume fraction
  - CBD has 5% ionic conductivity of pure electrolyte (Zielke 2015)



Zielke (2015)



Volume fraction of CBD for dense and nanoporous treatments for various CBD loadings

Binder weight fraction	Dense volume binder:particle	Porous volume binder:particle
0.04	0.10	0.15
0.06	0.16	0.23
0.08	0.22	0.31
0.10	0.28	0.40

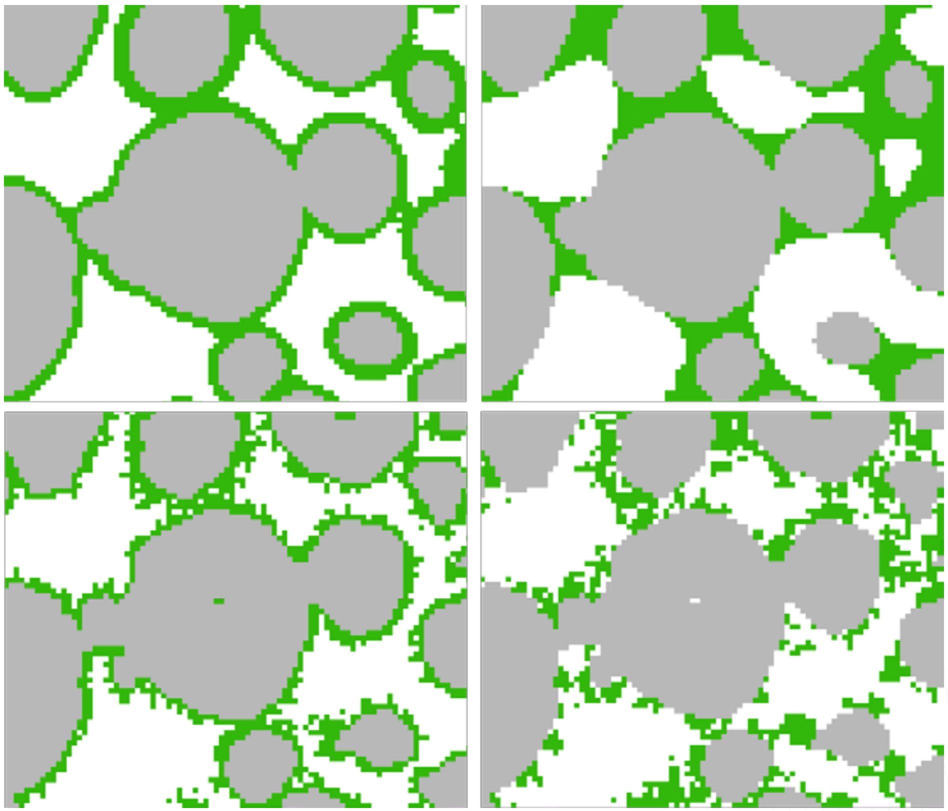
Sub-scale nanoporosity important to capture

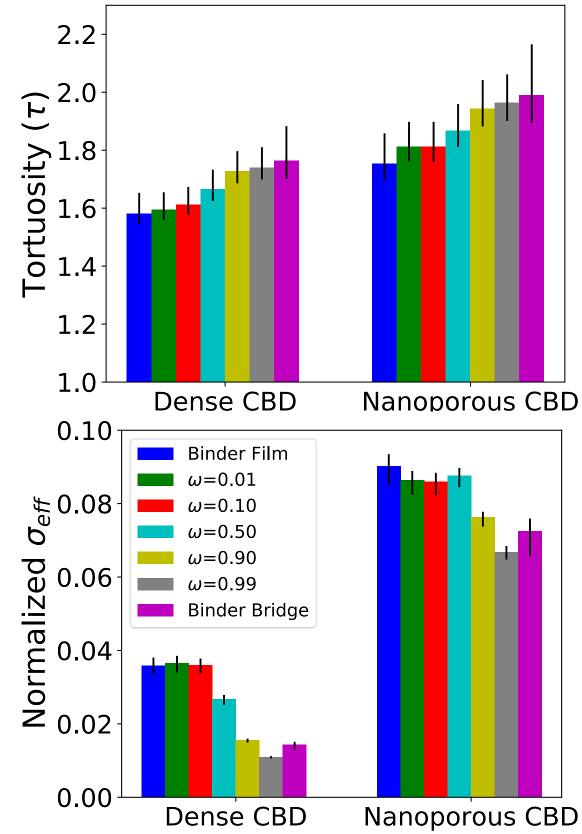
Technical: CBD Morphology Comparisons

	Coating	Bridge/Secondary

SNL
FEM
Voxelated

Purdue
FVM
Voxelated





Non-uniform binder:

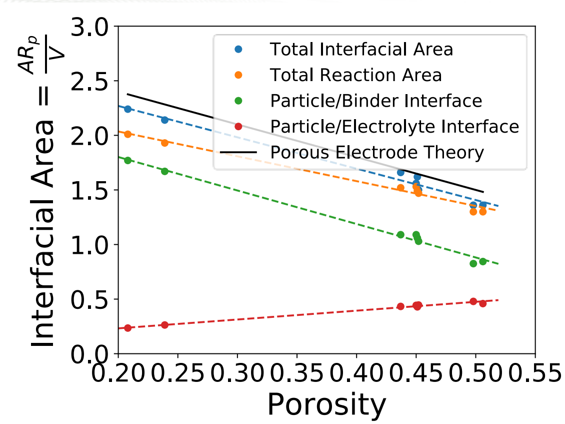
- Increases tortuosity
- Decreases conductivity

Nanoporosity:

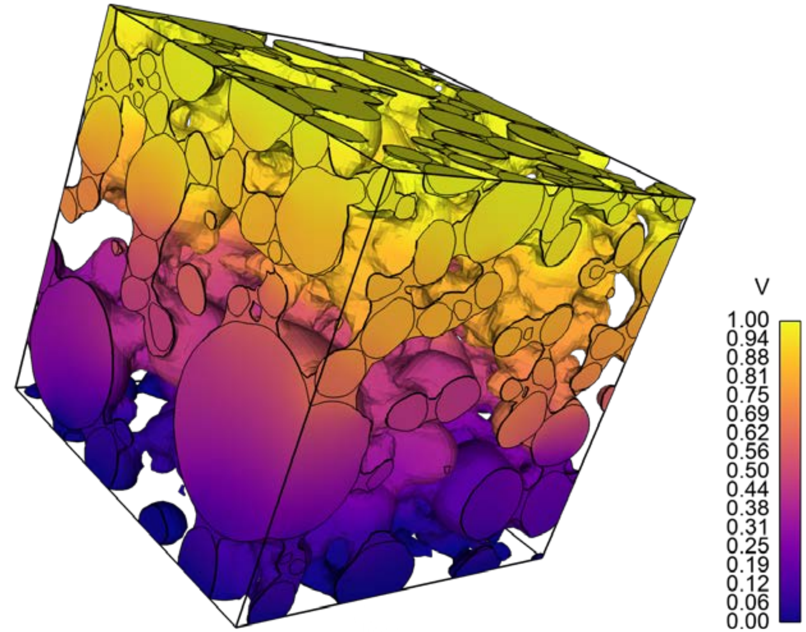
- Increases tortuosity
- Increases conductivity

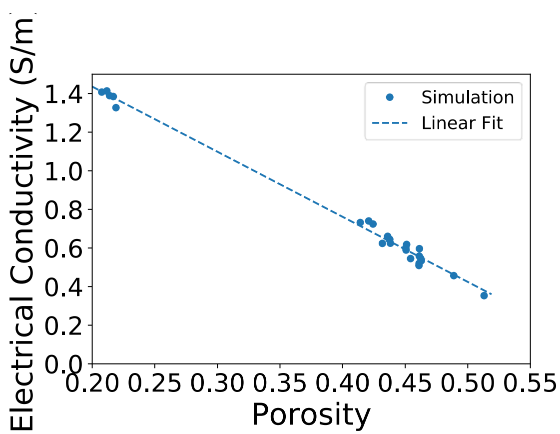
Comparisons between techniques and methods yield insightful results

Technical: Upscaling Mesoscale to Macroscale

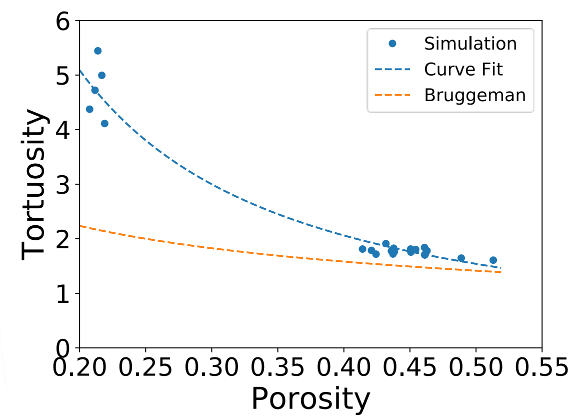


Total interfacial area follows trends of porous electrode theory





Electrical conductivity linear with porosity



Tortuosity values significantly exceed Bruggeman at low porosity

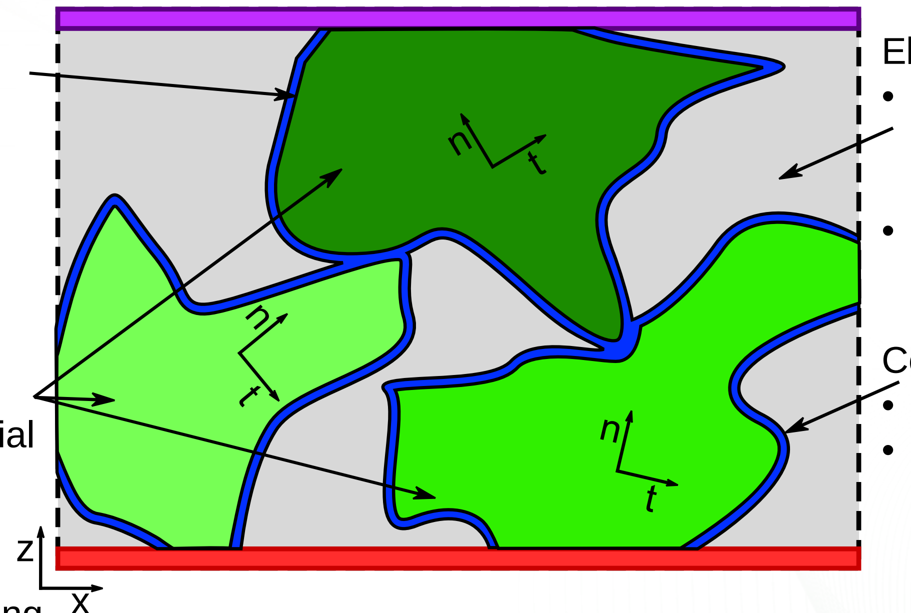
Mesoscale modeling can impact battery-scale abuse sims.

Ferraro et al, in preparation

Technical: Electrochemical-Mechanical Mesoscale Discharge Simulations

Particle Interface:
- Butler-Volmer reaction
- OCV from Smekens (2015)

Particles:
- Species – Li transport
  - Chemical potential
  - Stress potential
- Electrical – Ohm's law
- Mechanics - Elastic
  - Li-induced swelling

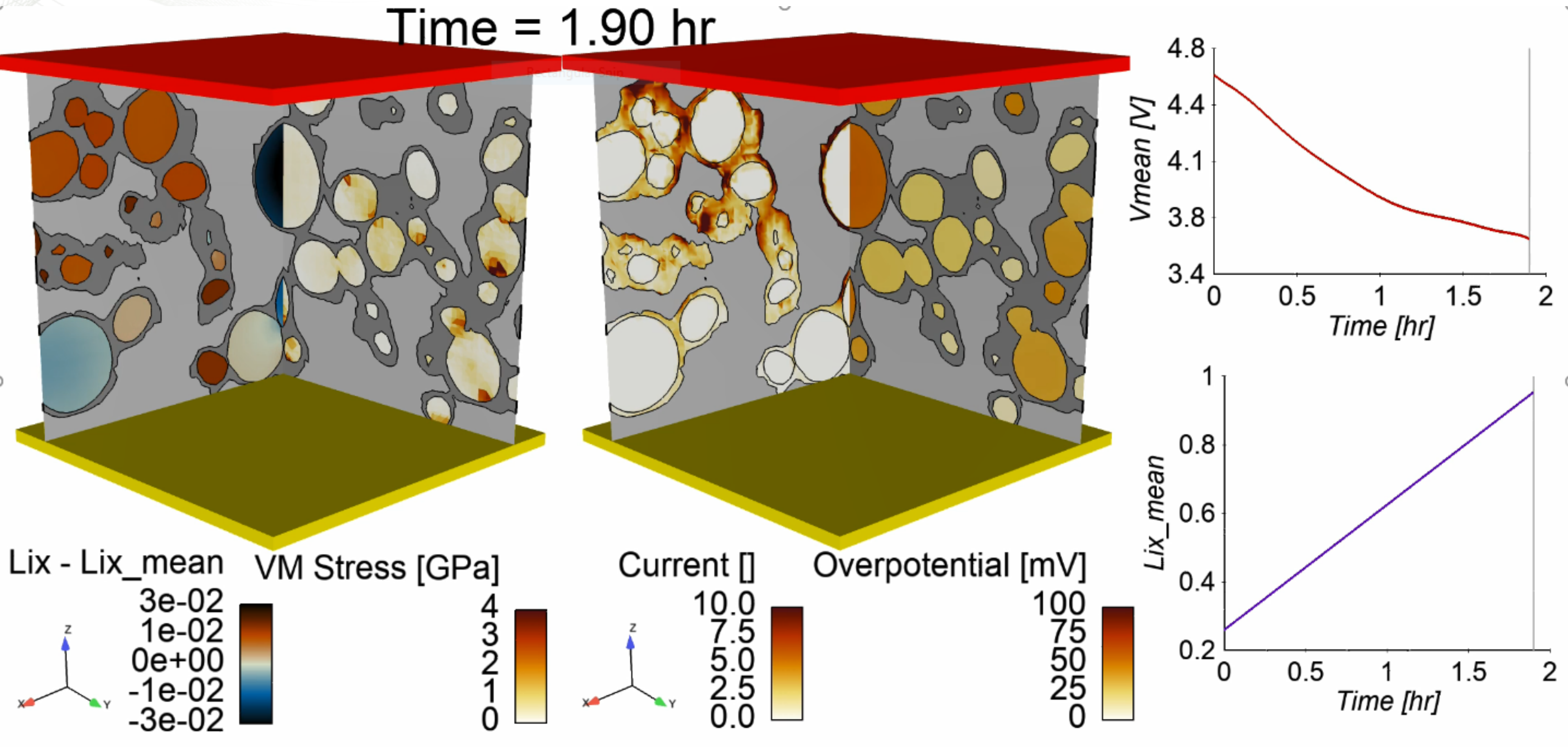


Electrolyte:
- Species – Li^+ transport
  - Nernst-Planck fluxes
  - Electroneutrality for PF_6^-
- Current conservation

Conductive binder:
- Species – Porous Li^+ transport
- Electrical
  - Solid: Porous Ohm's law
    - Strain-dependent electrical cond.
  - Liquid: Ionic conservation & electroneutrality
- Mechanics – Elastic

Mathematical formulation builds off of Mendoza (2016) LCO studies

Technical: Electrochemical-Mechanical Mesoscale Discharge Simulations



Predict discharge curve while interrogating mesoscale details

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Ferraro et al, in preparation

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Responses to Previous Year Reviewers' Comments

- Question regarding binder morphology inclusion of surface tension, source of properties (amorphous?)
  - Binder morphology approach was designed to mimic surface-tension-driven consolidation, but is not directly physics-based. Where available, properties were obtained from binder coatings manufactured in a similar fashion (e.g. Grillet (2016), Zielke (2015)).
- Thermal sensitivity of predicted properties
  - Effective property simulations presented in 2017 did not include temperature dependence. However current coupled electrochemical-mechanical-thermal simulations do include temperature-dependent properties and thermal expansion.

Any proposed future work is subject to change based on funding levels

Responses to Previous Year Reviewers' Comments

- How particles are tied together, nodal rigid body, damping?
  - While the particles are in tied contact, they are not constrained to rigid motion, but are elastic. The only constraint is that the contact area can not change. That said, the current formulation allows for soft binder to be inserted between particles, which introduces damping. Current investments include solutions involving finite deformation contact mechanics.

Any proposed future work is subject to change based on funding levels

Collaboration and Coordination with Other Institutions

Organization	Type	Relat.	VT?	Extent
Oak Ridge National Laboratory	Natl. Lab	Prime	Y	Upscaling to battery sims, experiments
Lawrence Berkeley National Laboratory	Natl. Lab	Peer Sub	Y	Tomography, microscale simulations, experiments
Argonne National Laboratory	Natl. Lab	Peer Sub	Y	Tomography
National Renewable Energy Laboratory	Natl. Lab	CAEBAT	Y	General collaboration, sharing of results and ideas
Purdue University	Acad.	CAEBAT	Y	Microstructure simulation collaboration
Duracell	Indust.	CRADA	N	Shared microstructure / electrochemistry development

Broad collaboration improves our work

Remaining Challenges and Barriers

- High-quality, controlled mesostructure reconstructions at controlled conditions that resolve active binder phase
  - Active binder apparently impossible to detect with X-rays
- Availability / quality of mesoscale validation data
  - Significant uncertainty in input parameters boosts importance of validating results against experimental data
- Computational requirements for electrochemistry simulations
  - Large domains approach 50 million elements and likely require 10,000 CPU core-hours, 10s of samples needed
- Mesostructure evolution needs finite deformation mechanics
  - Will become complex with binder added

Any proposed future work is subject to change based on funding levels

We address these risks in our future work

Proposed Future Research

- Discrete element method (DEM) simulations of entire manufacturing process to computationally derive mesostructure including particles + binder
  - Will be computationally expensive, but have expertise and resources
- Coordinate with ORNL/LBNL to measure electrode-scale properties for model validation
- Complete parametric studies of effective properties and electrochemical-mechanical performance for upscaling
- Further develop computational scalability and efficiency, pursue an exascale multi-scale coupling approach
- Develop multi-code coupling for finite deformation Lagrangian contact mechanics combined with Eulerian transport and electrochemistry

Any proposed future work is subject to change based on funding levels

Future work tailored to address key risks, milestones

Summary

- **Objective:** Create high-fidelity microstructure simulations of Li-ion battery electrodes to inform battery-scale simulations of operation and abuse
- **Results:**
  - Demonstrated robust and verified approach for three-phase cathode mesostructure representation
  - Developed understanding for the role of nanoporous carbon binder domains and how to construct them
  - Upscaled mesoscale results for use in macroscale (ORNL) battery abuse code
- **Results (cont.):**
  - Demonstrated fully-coupled electrochemical-mechanical-thermal simulations of NMC half-cell discharge
- **Future work:**
  - Efficiently and accurately create three-phase mesostructures using DEM
  - Complete parametric studies of effective properties and electrochemical cycling performance for NMC half-cells
  - Integrate microstructure simulation capability into battery-scale simulation framework

Any proposed future work is subject to change based on funding levels

Technical Back-Up Slides